INVESTIGATION OF NATURALLY CONVECTIVE HEAT EXCHANGE IN THE PRESENCE OF A HETEROGENEOUS CATALYTIC REACTION

S. G. Chefanov and É. A. Shtessel

UDC 536.25

The intensity of heat transmission because of natural convection in the presence of a heterogeneous catalytic reaction is investigated experimentally taking account of temperature and concentration factors.

The intensity of heat transmission with thermal natural convection, which is characterized by the dependence of the Nusselt number $Nu$ on the Rayleigh number $Ra$, has been studied sufficiently well at present. At the same time, the appearance of a product with a molecular weight different from that of the initial substance is possible during the progress of a heterogeneous chemical reaction. This should result in a dependence of the density on the coordinates, i.e., concentration convection can appear under definite conditions. At the same time, if the product being obtained as a result of the reaction is heavier than the initial substance, then the dependence of the density on the concentration of the product will have a stabilizing effect in the presence of an external negative temperature gradient. It is hence interesting to investigate the dependence of the intensity of heat transmission on the natural convection described by two causes: thermal and concentration.

Experiments were performed on a model reaction. The reaction of oxidizing sulfur dioxide on a platinum catalyst was taken as such a reaction which is used in investigations of heat transmission because of thermal and concentration convection. The product obtained as a result of the reaction is heavier than the initial substance. It is known that the reaction of oxidizing sulfur dioxide proceeds exothermically with the thermal effect $Q = 22 \text{ kcal per mole of SO}_3$ [1].

The tests were conducted with an excess of $\text{SO}_2$. The mixture consisted of 87% $\text{SO}_2$ and 13% $\text{O}_2$. The reaction of oxidizing $\text{SO}_2$ proceeds in the kinetic domain in the 300-450°C temperature range [2]. Passage into the diffusion domain occurs in the 750-800°C temperature range, where the reverse reaction yields an essential contribution at these temperatures. Hence, the tests to study heat transmission were performed at 300-450°C temperatures.

Two problems are manifest in studying the progress of the reaction under natural convection conditions: first is the origination and development of convection because of the heterogeneous catalytic reaction; the second is the investigation of the influence of the development of convection on the progress of the reaction. The influence of the reaction on the intensity of convection is evidently realized in both the kinetic and diffusion modes. The reverse influence is most substantial just in the diffusion domain.

An electrothermographic method [3] was used to investigate the heat transmission. A platinum catalyst in the form of a 50-$\mu$-thick foil was used as measuring element — as a resistance thermometer. The foil had transverse slits to increase the initial resistance. The diagram of the measuring cell is shown in Fig. 1.

The platinum foil 1 was glued to a mica plate 2 of $\approx 100 \mu$ thickness, which was supported on the thin porcelain rods 3. The rods were fastened to a cylindrical vessel filled with the heat insulator 4. The platinum foil plate had a 64 mm diameter, equal to the diameter of the outer vessel. Because of this construction the reaction could only proceed on the upper surface of the foil. Together with the fastened platinum element, the inner vessel was placed in the outer vessel of 130 mm height. The end face of the outer vessel was the 0.5-mm-thick glass plate 6 with 100-200-$\mu$-diameter holes. The holes were arranged uniformly over the whole surface of the plate. The spacing between the holes was 2 mm. Such a plate has no diffusion resistance but is hydrodynamically impermeable to natural convection. Above a porous plate parallel to it the reactive mixture is

bubbled through at a definite velocity. The outer vessel was heated by the electric spiral, which accomplished preliminary heating of the measuring cell. After the preliminary heating, the electric circuit of the platinum resistance thermometer was switched in, and the platinum catalyst was heated to the required temperature. Uniformity of the heating of the platinum element was checked by special thermocouples (\( \approx 10 \mu \text{ thick} \)). The temperature fluctuations along its surface did not exceed \( \pm 1.5^\circ \text{C} \) for the maximum temperatures on the element. Therefore, convection could only occur over the platinum element. The spacing between the porous baffle and the catalyst could be varied. The temperature of the platinum catalyst was determined to no worse accuracy than 1% by its resistance. The temperature of the porous baffle and the side surface was found by the thermocouples 9. The platinum resistance thermometer was connected in the arm of the measuring bridge.

The resistance and the current flowing through the catalyst were measured in the test. The catalyst temperature and the electrical power \( N \) supplied to the catalyst were computed from these results.

A standard \( \text{SO}_2 + \text{N}_2 \) mixture, whose thermophysical properties are quite similar to the properties of the reacting mixture, was initially bubbled through the preheated cell during performance of the experiments.

After equilibrium had built up, the platinum plate was heated by the current. The coefficients of heat elimination through the endfaces and side surface of the vessel were determined by conducting tests with an inert mixture. Then the rate of the reaction heat evolution could be determined by bubbling through the \( \text{SO}_2 + \text{O}_2 \) mixture. Preliminary warming of the outer surface permitted increasing the initial resistance of the measuring element, but meanwhile decreasing the heat losses.

Since the main problem of the investigation was determination of the coefficient of heat transmission from a catalytic surface to an outer surface over which a fresh mixture is blown, then this heat flux should be extracted from the total heat losses from the element.

Let \( N \) be the electrical power extracted from the catalyst and let \( \alpha \) be the coefficient of heat elimination from the catalyst through the upper endface surface. Then the following expression can be written in the absence of a reaction:

\[
N = \alpha S (T_1 - T_0) + q(T_1),
\]

where \( q(T_1) \) denotes the total heat flux down through the side walls.

The expression (1) was used to determine the function \( q(T_1) \) when the outer surface was blown off by the mixture of sulfur dioxide and nitrogen. The platinum element was hence 4 mm from the upper surface. Convection is completely absent at this height, since the value of the Rayleigh number is less than critical for such a gap. Therefore, in this case the quantity \( \alpha \) can be computed by means of the formula \( \alpha = \frac{\lambda}{h} \). By knowing \( \alpha \), the desired function \( q(T_1) \) was determined from (1) by means of the measured \( N \) and \( T_1 \).

Upon replacement of the inert gas mixture by the reacting mixture, (1) will be

\[
N = \alpha S (T_1 - T_0) + q(T_1) + q(T_1),
\]

where \( q \) is the heat liberated because of the reaction. Comparing (1) and (2) at the same temperature (or, equivalently, at a constant electrical resistance), we find the rate of reaction heat liberation.